

Is $\text{U}_3\text{Ni}_3\text{Sn}_4$ best described as near a quantum critical point?

C. H. Booth,¹ L. Shlyk,² K. Nenkov,^{2,3} J. G. Huber,⁴ and L. E. De Long⁴

¹*Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

²*IFW, Institute for Solid State and Materials Research, POB 270016, 01171 Dresden, Germany*

³*International Laboratory of High Magnetic Fields and Low Temperatures, Gajowicka 95, 53-529, Wrocław, Poland*

⁴*Department of Physics and Astronomy, University of Kentucky, Lexington, KY 40506-0055, USA*

(Dated: Draft: February 14, 2003)

Although many non-Fermi liquid (NFL) materials are structurally or chemically disordered, the role of disorder remains unclear. In particular, a few systems have been discovered that may be stoichiometric and well ordered. To test whether $\text{U}_3\text{Ni}_3\text{Sn}_4$ belongs in this class, we present measurements of the x-ray absorption fine structure (XAFS) of polycrystalline and single crystalline $\text{U}_3\text{Ni}_3\text{Sn}_4$ that are consistent with no measurable local atomic disorder. We also present temperature-dependent specific heat data in applied magnetic fields as high as 8 T that show features that are inconsistent with the antiferromagnetic Griffiths' phase model, but do support the conclusion that a FL/NFL crossover temperature increases with applied field. These results are inconsistent with theoretical explanations that require strong disorder effects, but do support the view that $\text{U}_3\text{Ni}_3\text{Sn}_4$ is one of very few stoichiometric, ordered materials that exhibit non-Fermi liquid (NFL) behavior, and is best described as being near an antiferromagnetic quantum critical point.

PACS numbers: 72.15.Qm, 61.10.Ht, 71.23.-k, 71.27.+a

I. INTRODUCTION

$\text{U}_3\text{Ni}_3\text{Sn}_4$ displays characteristic non-Fermi liquid behavior (NFL). For instance, the low-temperature magnetic susceptibility χ diverges as $T^{-0.3}$, the Sommerfeld coefficient for the electronic term in the specific heat $\gamma = C_{\text{el}}/T$ diverges as $-T^{0.5}$, and the resistivity varies as $T^{1.79}$. Such behavior is at odds with the standard Fermi liquid (FL) description ($\chi \sim C_{\text{el}}/T \sim \text{const.}$, $\Delta\rho \sim T^2$) of Landau.¹ The present study attempts to differentiate between the applicability of various theoretical models describing NFL behavior in this system by searching for the presence of local lattice disorder and measuring how the electronic part of the specific heat is affected by applied magnetic fields.

Current models describing NFL behavior fall into a few general classes, including those that invoke close proximity to a zero-temperature phase transition, competition between interactions such as Ruderman-Kittel-Kasuya-Yosida (RKKY) and Kondo effects, single-impurity versus non-single-impurity models, and those that include magnetic-interaction disorder. For instance, non-Fermi liquid behavior in the high-temperature superconductors and in some of the heavy fermion systems has been postulated to be due to the proximity of these systems to a zero-temperature magnetic phase transition.² We will refer to such models as anti- or ferromagnetic quantum critical point (AF-QCP or FM-QCP) models. Indeed, a number of f -electron compounds and alloys have been described as near a QCP, based on the entry of a system into a magnetic phase with increasing applied magnetic field or pressure or via chemical substitution ("chemical pressure").³ Millis⁴ and others⁵⁻⁸ have developed the theory of critical fluctuations at temperatures above such a magnetic/non-magnetic QCP, building off earlier work

by Hertz.⁹ The proximity of a magnetic phase is not necessary to obtain an NFL state, however. For instance, a multichannel Kondo model¹⁰ also results in non-Fermi liquid behavior. In addition, even though these theories all use a single magnetic interaction strength between the f and the conducting electrons (as opposed to a distribution), the first-discovered NFL systems also contain some form of lattice disorder, usually in the form of chemical substitution. This raises the possibility that lattice disorder plays an important role in NFL physics. In fact, a broad distribution of effective moments has been observed in several systems (for instance, CeRhRuSi_2 ¹¹ and UPdCu_4 ¹²). These observations prompted researchers to consider the role of "magnetic interaction disorder" as a microscopic origin for non-Fermi liquid effects. One simple theory utilizes only Fermi liquid concepts with a distribution of Kondo interactions, and is known as the "Kondo disorder model" (KDM).^{12,13} Other models that consider disorder in the vicinity of a zero-temperature fixed point are known as Griffiths' phase models (note that the KDM is also a Griffiths' phase model, but is not usually classified as such). These include the Griffiths'-McCoy singularities that occur in a disordered Kondo system, but whose properties mainly derive from antiferromagnetic RKKY interactions within only a few local clusters.^{14,15} Another possible origin of a Griffiths' phase occurs when disorder-induced Anderson localization occurs in the vicinity of a metal-insulator transition.¹⁶ To clarify the discussion, we will refer to the former model as the antiferromagnetic Griffiths' phase, or AF-GP, and to the latter model as the metal-insulator-transition Griffiths' phase, or MIT-GP.

Although most existing NFL materials have some intrinsic disorder, a growing number of NFL materials have recently been identified that appear to be stoichiometric

and structurally well-ordered at ambient pressure. Some examples include YbRh_2Si_2 ,¹⁷ CeNi_2Ge_2 ,¹⁸ CeCoIn_5 ,¹⁹ and $\text{U}_3\text{Ni}_3\text{Sn}_4$.²⁰ Although some of the physical properties of these systems agree with those predicted by the QCP model proposed by Millis,⁴ none of these materials display properties that completely agree with it. In addition, it is difficult to uniquely differentiate between “pure” QCP models and Griffiths’ phase models, especially since the Griffiths’ models have critical exponents that depend upon the degree of disorder in a way that is presently impossible to quantitatively relate to experimental measures of disorder.²¹ Such is the case for $\text{U}_3\text{Ni}_3\text{Sn}_4$.

Careful consideration of the disorder-based and the pure QCP models involves comparisons both of electronic and magnetic properties to theory, and thorough characterization of the degree of structural and magnetic order of the samples. The $\text{U}_3\text{Ni}_3\text{Sn}_4$ situation is complicated by the fact that $\text{U}_3\text{Ni}_3\text{Sn}_4$ has been shown to have a Fermi liquid ground state below about 0.4-0.5 K, with NFL behavior occurring above this crossover region.²² Regardless, in the NFL region, the electrical resistivity goes as $\Delta\rho = \rho(T) - \rho_0 \sim T^{1.79}$.²⁰ This dependence is roughly consistent with the AF-QCP result of $\Delta\rho \sim T^{1.5}$, if one takes into account the possibility of local “hot spots” of strong quasiparticle scattering at particular points in reciprocal space.⁸ The experimental magnetic susceptibility χ diverges as $T^{-0.3}$, at odds with the predictions of the non-divergent AF-QCP result of $T^{1.5}$. The electronic part of the specific heat diverges as $C_{\text{el}}/T \sim -T^{0.5}$, consistent with an AF-QCP. Alternatively, these results can be self-consistently explained with a AF-GP phase^{14,15} and a critical exponent of $\lambda = 0.7$, which produces comparably good fits to the data.²² In addition, these data are qualitatively consistent with the two-channel Kondo model^{23–26}, although fits using this model require a very high spin-fluctuation energy.²⁰ Comparisons to the KDM are only qualitative as well, since the KDM predicts logarithmic divergences of the magnetic susceptibility and the specific heat and a linear temperature dependence of the electrical resistivity, all of which are clearly at odds with the experimental data. In addition, measurements at applied pressures up to 1.8 GPa²⁷ indicate that the low-temperature FL ground state of $\text{U}_3\text{Ni}_3\text{Sn}_4$ extends to higher temperatures with increasing pressure. A scaling analysis of the FL/NFL crossover temperature as a function of applied pressure strongly implying a negative-pressure magnetic QCP, which has been estimated at -0.04 ± 0.04 GPa.

Although the measured properties of $\text{U}_3\text{Ni}_3\text{Sn}_4$ do not clearly support any of the various NFL models, there is little evidence to suggest that any disorder exists in this system, apparently not supporting either the KDM or the Griffiths’ phase models in spite of their consistency with thermal and magnetic data. In particular, single crystals of the material form, and x-ray diffraction studies of these crystals show the material to be consistent with the nominal stoichiometry. Moreover, the residual

resistivity is as low as $7 \mu\Omega\cdot\text{cm}$ ²⁰. Nevertheless, some forms of lattice disorder can be difficult to detect using standard diffraction techniques. For instance, if disorder occurs in a non-periodic fashion, such as in glassy regions or very small domains, only a local structural probe such as x-ray absorption fine-structure (XAFS) or pair-distribution function (PDF) analysis of powder diffraction data will be sensitive to it. In addition, no temperature dependent structural studies have been performed, and disorder broadening of the displacement parameters (Debye-Waller factors) can easily be confused with large vibrational amplitudes. Therefore, we have undertaken a direct test of the degree of structural order in $\text{U}_3\text{Ni}_3\text{Sn}_4$, using XAFS spectroscopy. Although the crystal structure of $\text{U}_3\text{Ni}_3\text{Sn}_4$ limits the accuracy of the estimated maximal disorder levels as detailed below, our measurements are consistent with no disorder within experimental error, for both single crystals and polycrystals, based on temperature dependent data from all three investigated absorption edges.

As implied above, merely having the lattice be (measurably) well ordered may not rule out magnetic interaction disorder. Since the Griffiths’ phase models only require rare clusters of competing interactions to form, one can only place upper limits on the possible sizes of such clusters experimentally. In addition, some heretofore more subtle Kondo disorder mechanism may still be applicable (for instance, if large fluctuations in the conduction density of states accompanies fluctuations in the hybridization strength in the presence of lattice disorder²⁸). In any case, the AF-GP model makes quantitative predictions regarding the magnetic field dependence of certain properties, including the specific heat. Below we compare such measurements to the AF-GP predictions and find they are inconsistent. Instead, we find these data are more consistent with the presence of a low-lying magnetic phase. This analysis is similar, but with contrasting results to that done on the $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$.²⁹

The rest of this paper is organized as follows. The XAFS measurements are described in Sec. II, including a description of the sample preparation and the technique. Sec. III relates the results from the measurements of specific heat as a function of temperature and applied field. These results are discussed in relation to the various NFL theories in Sec. IV, and the final conclusions are summarized in Sec. V.

II. XAFS MEASUREMENTS

A. Background

$\text{U}_3\text{Ni}_3\text{Sn}_4$ crystallizes into a *bcc* structure, in the $I\bar{4}3d$ space group with the lattice parameter 9.3524 Å and a position *x* parameter value of 0.082 describing the Sn displacements (Fig. 1).²⁰ Crystallographically, the structure is fairly simple, with a single site for each atomic species. The local environments are rather more compli-

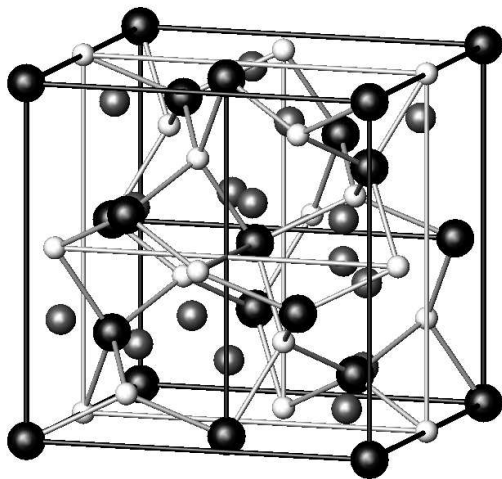


FIG. 1: Crystal structure of $\text{U}_3\text{Ni}_3\text{Sn}_4$. Black balls represent uranium, gray tin and white nickel. The materials is bcc with space group $I43d$ and $a=9.3524$ Å.

cated, although the nearest-neighbor shells are fairly well separated. For instance, U has 4 nearest-neighbor Ni's at 2.86 Å, followed by 8 Sn neighbors at 3.24 Å. Ni has 4 Sn neighbors at 2.61 Å and 4 U neighbors at 2.86 Å. Sn has 3 Ni neighbors at 2.61 Å, followed by 6 U neighbors at 3.24 Å and 3 Sn neighbors at 3.50 Å. (Fig. 2).

B. Experimental

Three of the samples are polycrystalline with nominal stoichiometries $\text{U}_{3.0}\text{Ni}_{3.0}\text{Sn}_{4.0}$, $\text{U}_{2.9}\text{Ni}_{3.0}\text{Sn}_{3.9}$, and $\text{U}_{3.0}\text{Ni}_{3.1}\text{Sn}_{3.9}$. Two single-crystal samples were also measured with nominal stoichiometries of $\text{U}_{3.0}\text{Ni}_{3.0}\text{Sn}_{4.0}$ and $\text{U}_{2.9}\text{Ni}_{3.0}\text{Sn}_{3.9}$, and are the same samples as those reported in Ref. 20. All XAFS data were collected on beam lines 4-1 and 4-3 at the Stanford Synchrotron Radiation Laboratory (SSRL) using half-tuned Si(220) double monochromator crystals. Samples were placed into a LHe flow cryostat. Polycrystalline samples were ground into a fine powder under acetone, passed through a 40 μm sieve and brushed onto scotch tape, with stacked layers corresponding to a change of one absorption length at each measured edge. Data for the polycrystals were collected at various temperature between 20 K and 300 K at the U L_{III} , and Sn K edges in transmission mode, and at the Ni K edge in fluorescence mode using a 4-pixel Ge detector.³⁰ Single crystal data were collected at 20 K at the U L_{III} and the Sn and Ni K edges in fluorescence mode. Several scans were obtained for each sample at each edge and temperature, and were fit separately to crosscheck the error estimates. Dead-time and self-absorption³¹ corrections were applied to the fluorescence data.

Data were reduced and fit in r -space using the RSXAP package.³²⁻³⁴ In particular, the XAFS function $\chi(k)$ is

defined as $\mu(k)/\mu_E(k) - 1$, where $\mu(k)$ is the absorption coefficient as a function of the photoelectron wave vector k , and $\mu_E(k)$ is the so-called “embedded atom” background absorption that is proportional to the number of generated photoelectrons. The wave vector is defined as $k = \sqrt{\frac{2m_e}{\hbar^2}(E - E_0)}$, and the photoelectron threshold energy is defined arbitrarily as the energy at the half-height of the edge, and is allowed to vary in subsequent fits. Examples of the $k\chi(k)$ data are shown in Fig. 3 for the polycrystalline samples. Data on single crystals is similar, both in quality and quantity.

The scattering amplitudes are all fixed to NS_0^2 , where N is the nominal number of neighbors in a given shell for the stoichiometric compound, and S_0^2 is the XAFS amplitude scale factor. Each data set was fit with a single value of S_0^2 , assuming full nominal site occupancies. All scattering paths also share a single value of E_0 . In the case where multiple temperatures were collected, average values of S_0^2 and E_0 were obtained and then held fixed for all temperatures for a given edge. Fixing N , S_0^2 and E_0 in the final fits drastically reduces the number of fit parameters, but assigns all the sources of lattice disorder into either the measured bond lengths, R , or the Debye-Waller factors, σ . In particular, the effect of vacancies

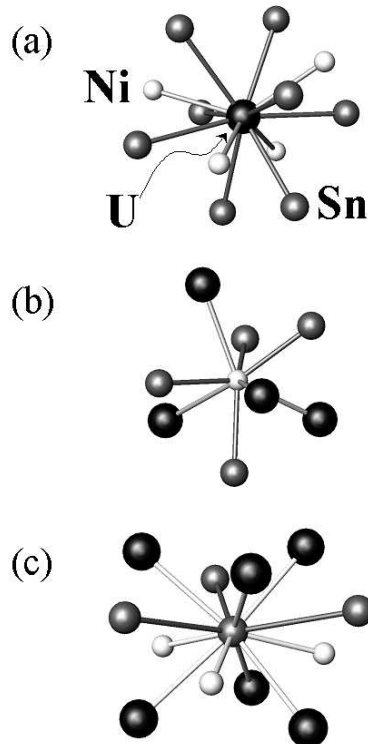


FIG. 2: Local crystal structure from the (a) uranium, (b) nickel, and (c) tin sites. See Sec. II A for details.

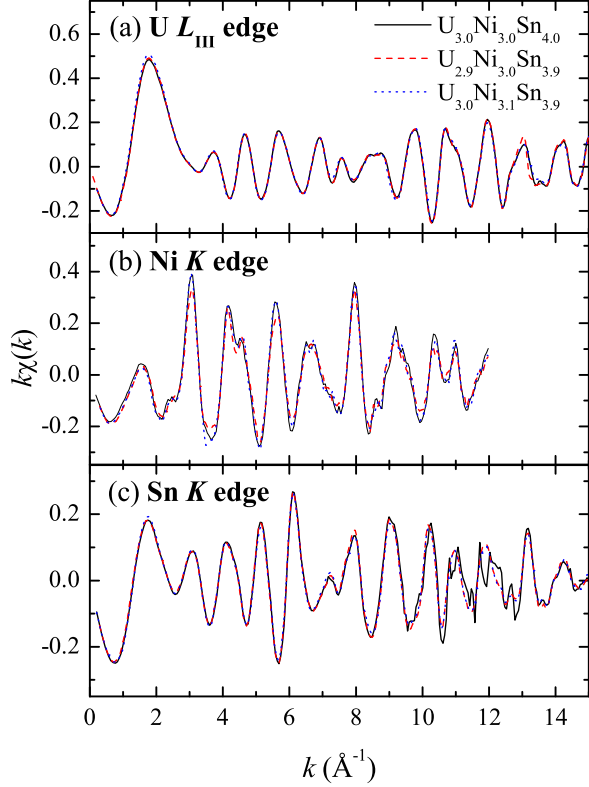


FIG. 3: XAFS data for the three polycrystalline samples. Single crystal data is quantitatively similar. Data from the various samples is very similar, and so is difficult to differentiate in the plot.

is placed on the Debye-Waller factors. Data on the single crystals were fit with different S_0^2 and E_0 than the polycrystalline data, although no major differences were observed. Reported error estimates use the larger of either a Monte Carlo estimate of the 1-standard deviation displacements (essentially equivalent to a covariance matrix without having to assume that the statistical- χ^2 is quadratic near its minimum), or the distribution of parameters obtained by fitting the individual scans at each temperature. Reported errors are generally consistent with those obtained from standard compounds, typically ± 0.005 Å in pair distance and ± 10 -20% in σ^2 for near neighbors, with the error roughly doubling after about 3 Å.³³

C. Results

Figures 4 and 5 show the Fourier transforms (FT) of $k^3\chi(k)$. Peaks in the FT's correspond to pairs in the local structure, however, it must be noted that constructive/destructive interference can occur (the functions are complex), the scattering profiles are not Gaussian, and phase shifts occur that place the peaks at distances in the

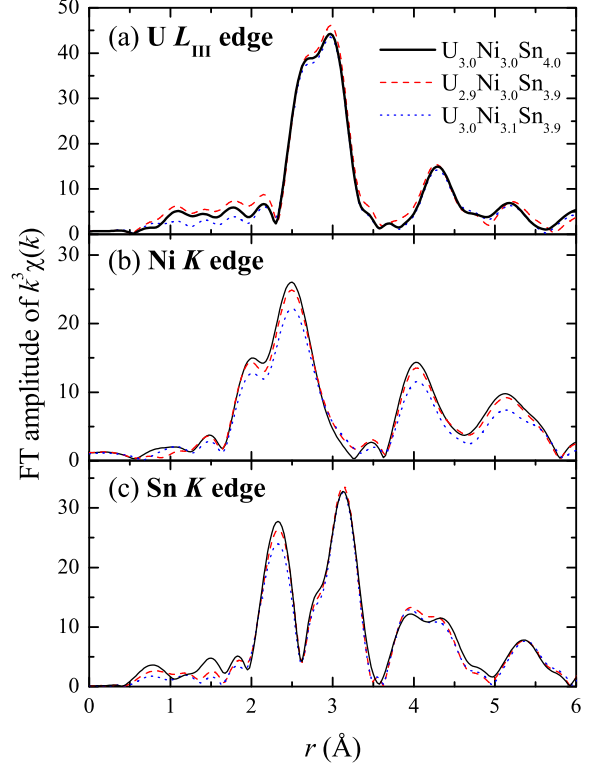


FIG. 4: Fourier transforms of the $k^3\chi(k)$ XAFS data for the powder samples. U and Sn (transmission) transforms are from $k=3.0$ - 15 Å⁻¹, while the Ni (fluorescence) transform is from 2.5 - 12.0 Å⁻¹, all Gaussian narrowed by 0.3 Å⁻¹.

FT's that are shorter than in the structure by an amount related to the species of absorber and backscatterer. Transmission data were collected out to a $k_{\text{max}} = 15$ Å⁻¹, and fluorescence data were collected out to a $k_{\text{max}} = 13$ Å⁻¹. Fig. 5 shows a comparison between the single and polycrystalline data for the $\text{U}_{3.0}\text{Ni}_{3.0}\text{Sn}_{4.0}$ samples using the same transform ranges.

There are visible differences between the various data sets for a given edge. In particular, the Ni and Sn edge data on the polycrystalline $\text{U}_{3.0}\text{Ni}_{3.1}\text{Sn}_{3.9}$ sample consistently show a reduced amplitude compared to $\text{U}_{3.0}\text{Ni}_{3.0}\text{Sn}_{4.0}$ at all temperatures, consistent with some disorder or the presence of an amorphous phase containing those elements. Although these differences are above the signal to noise, it is not possible to discern the exact cause of the differences from the fit results listed below. Differences between the polycrystalline and the single crystal data (Fig. 5) are similar in magnitude, but complications with analyzing single crystal data, such as dead-time and self-absorption corrections, are very likely the cause.

The basic procedure used here for searching for lattice disorder is to carry out fits assuming the nominal structure, then examine certain parameters for signs of disorder. In the fits, each distinct scattering shell in the

TABLE I: Final fit parameters to the U L_{III} , Ni K and Sn K edge data at 20 K on three polycrystalline samples of $U_3Ni_3Sn_4$ with various nominal stoichiometries. U L_{III} edge fits have $S_0^2 = 0.73 \pm 0.06$ and $\Delta E_0 = -10.3 \pm 0.4$. Ni K edge fits are from 2.5 to 13.0 \AA^{-1} (Gaussian narrowed by 0.3 \AA^{-1}) and from 1.4 to 5.0 \AA , and have $S_0^2 = 0.64 \pm 0.04$ and $\Delta E_0 = -0.5 \pm 0.5$. Sn K edge fits have $S_0^2 = 0.95 \pm 0.06$ and $\Delta E_0 = -8.3 \pm 0.1$. Diffraction data is provided for comparison (R_{diff}) and is from Ref. 20 on a single crystal sample of $U_3Ni_3Sn_4$ collected at room temperature.

pair	N	R_{diff}	R	$U_3Ni_3Sn_4$			R	$U_{2.9}Ni_{3.0}Sn_{3.9}$			R	$U_{3.0}Ni_{3.1}Sn_{3.9}$		
				σ^2	σ_{static}^2	Θ_{CD}		σ^2	σ_{static}^2	Θ_{CD}		σ^2	σ_{static}^2	Θ_{CD}
U-Ni	4	2.864	2.848(3)	0.0018(2)	-0.0009(4)	252(5)	2.848(4)	0.0019(4)	-0.0004(5)	282(2)	2.848(3)	0.0019(2)	-0.0005(5)	259(4)
U-Sn	8	3.237	3.228(2)	0.0011(2)	-0.0006(3)	241(1)	3.226(4)	0.0009(2)	-0.0009(3)	231(1)	3.226(3)	0.0009(2)	-0.0007(3)	233(1)
U-U	8	4.374	4.36(1)	0.0016(3)	-0.0005(3)	164(6)	4.355(5)	0.0014(2)	-0.0005(3)	173(2)	4.355(3)	0.0014(2)	-0.0000(3)	169(3)
U-Ni	2	4.676	4.67(1)	0.0015(3)			4.67(1)	0.0022(6)			4.67(1)	0.0022(4)		
Sn-Ni	3	2.609	2.604(3)	0.003(1)	0.001(1)	420(15)	2.597(3)	0.0027(2)	-0.0003(2)	349(4)	2.599(3)	0.0027(2)	0.0008(3)	359(3)
Sn-U	6	3.237	3.223(5)	0.0006(4)	-0.0017(5)	202(12)	3.232(7)	0.0016(2)	-0.0004(2)	246(2)	3.228(3)	0.0012(2)	-0.0004(2)	273(5)
Sn-Sn	3	3.497	3.50(3)	0.003(3)	-0.003(3)	172(17)	3.500(4)	0.004(1)	-0.000(1)	245(6)	3.496(3)	0.0017(5)	-0.0001(6)	250(20)
Sn-Sn	2	4.050	3.98(3)	0.003(3)			4.03(1)	0.01(1)			4.02(1)	0.0024(7)		
Sn-Ni	3	4.232	4.25(3)	0.002(1)			4.16(5)	0.01(1)			4.22(1)	0.005(2)		
Sn-Sn	6	4.594	4.60(3)	0.002(1)			4.598(4)	0.0034(3)			4.596(3)	0.0023(2)		
Ni-Sn	3	2.609	2.588(4)	0.0017(4)	-0.0004(5)	347(10)	2.585(6)	0.0014(3)	-0.0007(3)	334(10)	2.587(4)	0.0017(6)	0.0003(6)	353(4)
Ni-U	3	2.864	2.850(6)	0.0028(6)	0.0003(7)	252(7)	2.850(8)	0.0023(5)	-0.0005(5)	234(7)	2.853(6)	0.002(1)	-0.0005(9)	234(4)
Ni-Sn	3	4.231	4.24(1)	0.0005(9)			4.26(1)	0.000(2)			4.25(2)	0.004(3)		
Ni-Ni	3	4.374	4.35(1)	0.001(1)			4.35(1)	0.000(1)			4.37(1)	0.001(2)		
Ni-U	3	4.676	4.70(5)	0.005(7)			4.70(3)	0.003(4)			4.70(3)	0.003(2)		

nominal structure out to about 4.7 \AA is used at each edge. Fit results to the data from the polycrystalline samples are shown in Table I, and are compared to results inferred from diffraction measurements. Fit quality is very high; examples of the fit quality are shown in Fig. 6. Although all the polycrystalline data were collected as a function of temperature, we only show the fit results for the coldest measured temperature. No significant changes in the fit parameters occur with temperature, except that the Debye-Waller factors increase in a manner consistent with the correlated-Debye model³⁵ plus a temperature-independent offset σ_{static}^2 . Such off-sets can be used as indicators for non-thermal disorder (a prime example occurs in the colossal magnetoresistance manganese perovskites³⁶), but we see no evidence for σ_{static}^2 values inconsistent with zero disorder. Results for the correlated-Debye temperature Θ_{CD} and σ_{static}^2 are shown in Table I. Note that the preponderance of small, unphysically negative measurements of σ_{static}^2 are likely due to small underestimates of S_0^2 .

Fit results to the single crystal data were found to be the same as results from the polycrystalline data within the error estimates, and so are not reported here. No evidence for lattice disorder is observed, as exemplified by the consistently low values of the nearest-neighbor σ^2 's for the low temperature fits.

Finally, we consider the possibility of site interchange, or site/anti-site disorder. This possibility is very remote, however, given the big differences in the radii of the atoms involved, except for Sn/U interchange (covalent radii are 1.42 \AA , 1.15 \AA , and 1.41 \AA for U, Ni and Sn, respectively). Unfortunately, fitting the Sn and U XAFS data including some U/Sn interchange gives only a broad result: $s = 9 \pm 10\%$, where s is the percentage of Sn sitting on U (12a) sites. Fits including U/Ni and Sn/Ni site interchange were similarly imprecise. The principle difficulty in using XAFS to measure s between, for instance, U and Sn, is

that the local nearest-neighbor environments each have pair distances near $\sim 2.7 \text{\AA}$ and then near 3.2 \AA ; that is, no new peaks would appear in these spectra if site interchange occurs. This situation is in contrast to that in the UPdCu₄ system.³⁷ In any case, the single crystal diffraction results²⁰ should not have produced such high quality fits if much more than 5% of such interchange occurs.

As a further check for possible U/Sn interchange, we considered the possibility of Sn/U solid solutions in the $U_3Ni_3Sn_4$ phase. Since thermodynamic experimental information on the U-Ni-Sn ternaries is unavailable, we investigated the formation of Sn/U solid solutions by growing $U_3Ni_3Sn_4$ single crystals with 5wt% of Sn excess. X-ray diffraction studies of the solidified melt confirmed the presence of the $U_3Ni_3Sn_4$ phase without an additional formation of solid solution of U in Sn. This result led us to conclude that Sn/U site interchange is unlikely in $U_3Ni_3Sn_4$ phase.

III. FIELD-DEPENDENT SPECIFIC HEAT MEASUREMENTS

The specific heat of a $U_3Ni_3Sn_4$ single crystal from the same batch as the XAFS sample was measured between 1.8 and 30 K in applied magnetic fields up to 8 T using a Quantum Design PPMS. The specific heat data are shown in Fig. 7, plotted as C_{el}/T versus $T^{0.5}$. Here we have already subtracted the hyperfine and lattice contribution according to the specific heat data of $U_3Ni_3Sn_4$ reported previously.²² In zero applied magnetic field the data follow the $C_{\text{el}}/T \propto T^{0.5}$ behavior below 6 K indicative of the non-Fermi liquid regime. Increasing the applied magnetic field progressively depresses the specific heat so that C_{el}/T shows a deviation from square root behavior at lower temperatures. It is

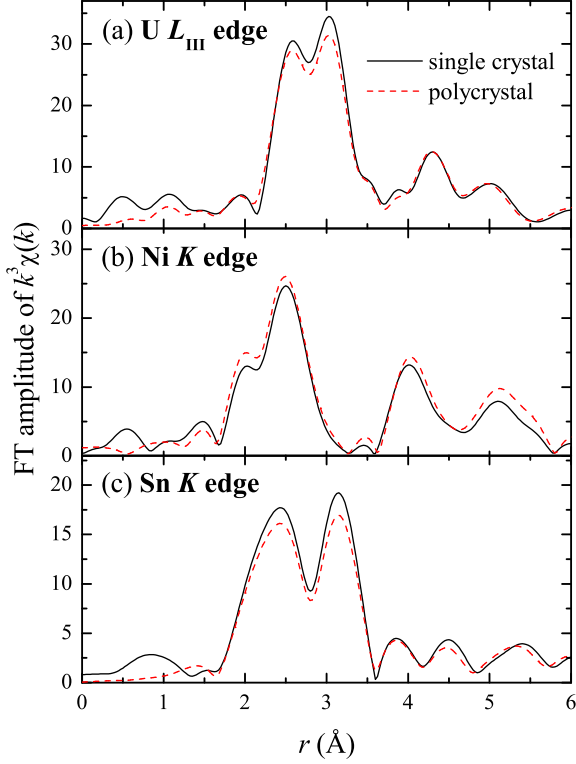


FIG. 5: Fourier transforms of the $k^3\chi(k)$ XAFS data comparing the polycrystalline and the single crystal data for the nominally $\text{U}_3\text{Ni}_3\text{Sn}_4$ samples with consistent transform ranges. U and Sn transforms are from $k=3.0\text{--}13 \text{ \AA}^{-1}$ while the Ni transform is from $2.5\text{--}12.0 \text{ \AA}^{-1}$, all Gaussian narrowed by 0.3 \AA^{-1} .

expected that C_{el}/T tends toward a constant value at temperatures lower than 1.8 K, suggesting the onset of a Fermi-liquid regime. These results strongly imply that applied fields take the system further away from an antiferromagnetic critical point. However, if one plots the data as $C_{\text{el}}(H, T)/C_{\text{el}}(H = 0, T)$ (Fig. 8), one might interpret the changes in applied field as arising from a new feature developing, Schottky-like in appearance. This observation leads to an alternative explanation of these data provided by the AF-GP model. In the high-field limit of this model, the specific heat should go as $C_{\text{el}}/T = AH^{2+\lambda/2}/T^{3-\lambda/2}e^{-\mu_{\text{eff}}H/T}$, where $\lambda = 0.7$ is the aforementioned critical exponent from the low temperature $C_{\text{el}}(H = 0)/T$ and the magnetic susceptibility data, μ_{eff} is an average effective moment of the antiferromagnetic clusters, and A is a constant that is difficult to calculate in the theory and is thus taken as arbitrary. This function predicts a Schottky-like anomaly near 3 K that is much larger than anything observed in the data, in contrast to that observed in the $\text{Ce}_{0.05}\text{La}_{0.95}\text{RhIn}_5$ data in Ref. 29. However, this analysis leaves open the question of the role of A and whether we are really in the high-field limit.

Instead, consider the form for the specific heat at any

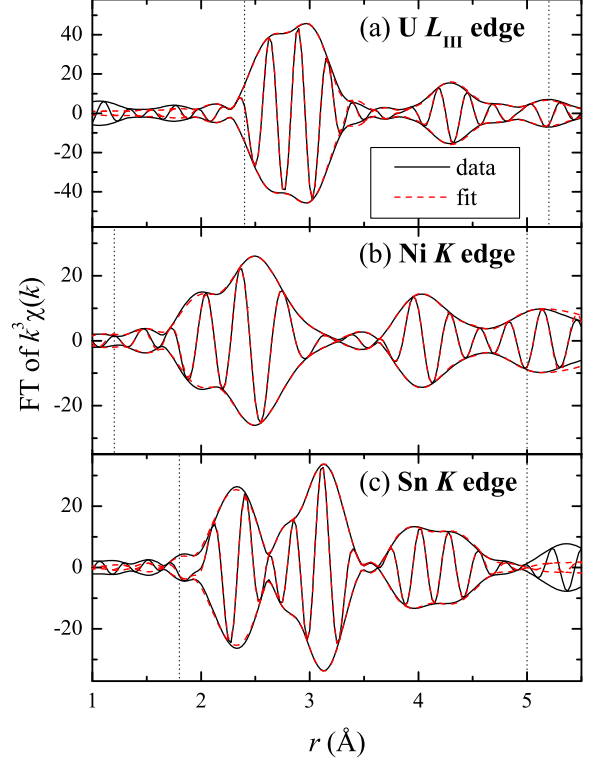


FIG. 6: Examples of the fits to the (a) $\text{U } L_{\text{III}}$ and the (b) $\text{Sn } K$ polycrystalline data. Each transform is represented by three lines. The inner oscillating line is the real part of the complex transform, while the envelope lines are \pm the amplitude of the transform. Vertical dotted lines show the r -space fit range. Transform ranges are as in Fig. 4.

field given in Ref. 15:

$$C_{\text{el}}(H, T) \propto \beta^2 \int_0^{\omega_0} d\Delta \Delta^{1-\lambda} (E_H^2 + \Delta^2) \times \text{sech}^2(\beta \sqrt{E_H^2 + \Delta^2}) \left[\ln \frac{\omega_0}{\Delta} \right]^{1-\theta}, \quad (1)$$

where Δ is the cluster tunneling energy, ω_0 is the tunneling energy for a single atom, β is $1/k_{\text{B}}T$, and θ is the percolation exponent. E_H is the magnetic energy of a given cluster, and is given by:

$$E_H(\Delta) = q\mu_{\text{B}} \left[\frac{1}{\gamma} \ln \left(\frac{\omega_0}{\Delta} \right) \right]^\phi H, \quad (2)$$

where q gives the magnitude of the average moment within a cluster, and $\phi = 1(1/2)$ for ferromagnets (antiferromagnets and spin glasses). For our data, $\lambda \approx 0.7$, the percolation scaling exponent for three dimensions with no magnetic order is $\theta = 3/2$, and $\phi = 1/2$. The tunneling frequency cutoff ω_0 is taken as one of only two fitting variables, with the other being q/γ^ϕ , which is taken as a single variable in what follows.

To avoid having to understand the proportionality constant in Eq. 1 we calculate the ratio $C_{\text{el}}(H, T)/C_{\text{el}}(H =$

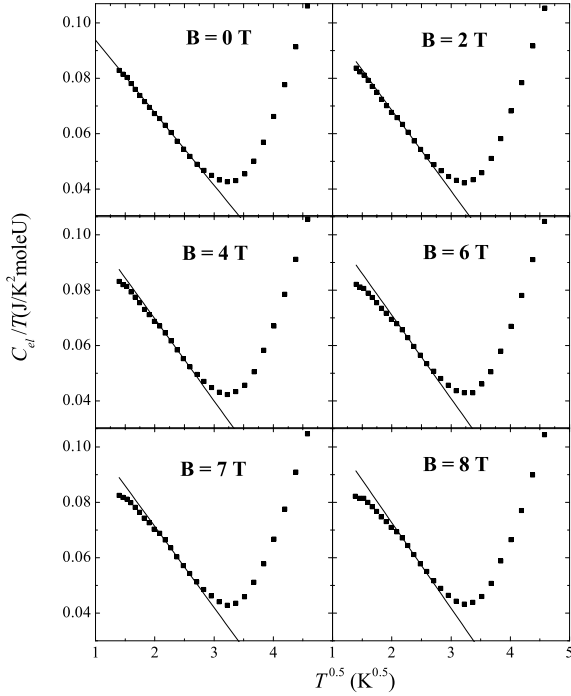


FIG. 7: C_{el}/T as a function of $T^{0.5}$ of $U_3Ni_3Sn_4$ single crystal for $B = 0, 2, 4, 6, 7, 8$ T. The straight line is a guide to the eye for the $C_{el}/T \sim T^{0.5}$ behavior.

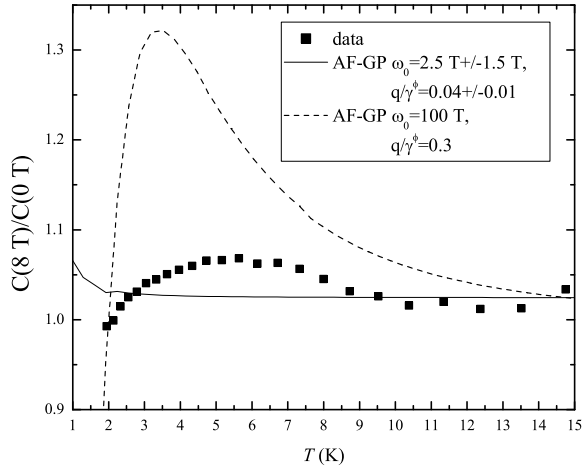


Fig. 8, Booth et al.

FIG. 8: The ratio of $C(H = 8T, T)/C(H = 0T, T)$ from Fig. 7. Lines are fits to the AF-GP model described in the text.

0, T). We find that no combination of the fitting variables produces a satisfactory fit. We show a typical fit attempt in Fig. 8, which has a hump in $C_{el}(H = 8T, T)/C_{el}(H = 0, T)$ in the vicinity of the observed hump. This “fit” produces far too large a hump, similarly to the high-field analysis above. The best fit actually places the Schottky-like anomaly below the observed range with

a very small effective moment ($\sim 1/100^{\text{th}}$ of that from $Ce_{0.05}La_{0.95}RhIn_5$). We therefore conclude that the AF-GP model (as posed) does not describe the physics in $U_3Ni_3Sn_4$.

IV. DISCUSSION

Deviations from the nominal structure in the fit results can occur in a number of ways. First, the measured S_0^2 amplitude reduction factors should be in a range that has been experimentally measured before, since this factor represents inelastic losses and errors in the theoretical backscattering amplitudes that only weakly depend on an individual system. Indeed, our measurements fall within acceptable ranges.^{33,37} Second, the temperature dependence of the Debye-Waller factors can be compared to a correlated-Debye model, with large offsets indicative of static (i.e. non-thermal) disorder or distortions. In all cases, we see no abnormally large offsets. Third, the measured pair distances should be reasonably close to those measured by diffraction, which is consistent with our measurements (Table I). Fourth, various site-interchange possibilities should be considered, such as U sitting on the (nominally) Sn (16c) site. Although these fits are not particularly sensitive to such interchanges, our measurements are consistent with no site interchange. Finally, the results from the single crystal and the polycrystalline samples are virtually identical. Together with the single crystal diffraction data and the absence of U/Sn solid solution phases in a melt with excess Sn, we must conclude that the $U_3Ni_3Sn_4$ system is structurally well ordered, and is much more ordered than, say, the $UPdCu_4$ system.^{36,38} Although such crystalline order does not rule out some other source of magnetic-interaction disorder that might be consistent with a Kondo disorder model or a Griffiths phase model, it certainly rules out a likely source. In addition, although these structural studies cannot rule out the presence of small amounts of disorder, previous work within the simplest form of the Kondo disorder model has shown that significantly more disorder would have to be present for that model to work (in $UPdCu_4$, for instance, Ref. 38 estimates that at least 0.002 \AA^2 of static disorder are necessary to produce NFL behavior from the KDM).

Even with small, undetected amounts of disorder, Griffiths’ phase models may be capable of describing the physics in this system. In the AF-GP model, for instance, the exact relationship between the required degree of disorder as measured by the number and distribution of antiferromagnetic clusters and the physical properties remains unclear. However, the AF-GP model makes very clear predictions about the evolution of the specific heat with applied magnetic field that we show in Sec. III do not describe the observed features. Until more precise, measurable predictions relating disorder to physical properties are obtained, we conclude that disorder presents at best a small perturbation to this system, leaving the AF-

QCP theory as the only current alternative.

There are other requirements for an AF-QCP theory, of course. For one, the system must be very near a magnetic/non-magnetic instability. Measurements under applied pressure indicate this instability may exist at a small negative pressure, based on a scaling of the resistivity behavior. We thus expect that if the magnetic phase is antiferromagnetic, an applied magnetic field will also move the system toward a Fermi liquid regime. The data presented in Fig. 7 indicate this system behaves exactly as one expects if the system is near a magnetic/non-magnetic instability, and are therefore consistent with models that include antiferromagnetism as a competing interaction.

V. CONCLUSION

In summary, we have measured the local structure around the constituent atoms in $\text{U}_3\text{Ni}_3\text{Sn}_4$ single and poly crystals as a function of temperature and stoichiometry. These data follow typical Debye model dependences in the measured pair-distance distribution widths with no (non-thermal) offsets, as well as agreeing well with the average structure given by previous single crystal diffraction studies.²⁰ In summary, the relatively low residual resistivity and all the structural evidence indicates that this system is structurally well ordered. We also report spe-

cific heat data that are clearly inconsistent with the antiferromagnetic Griffiths' phase model. These data instead show a recovery of Fermi liquid behavior under modest applied magnetic fields, indicating a probable "negative field" quantum critical point, consistent with work under applied pressure that indicates a negative-pressure QCP. Taking all these results together, we have ruled out the simple form of the KDM and the AF-GP theories. In addition, the lack of measurable disorder does not favor other disorder-based theories, such as the MIT-GP. With no indications to the contrary, we must conclude that the best current description of $\text{U}_3\text{Ni}_3\text{Sn}_4$ is that of a system near an antiferromagnetic quantum critical point.

Acknowledgments

We thank E. D. Bauer, A. J. Millis, E. Miranda, A. H. Castro Neto and D. L. Cox for many useful discussions. This work was partially supported by the Director, Office of Science, Office of Basic Energy Sciences (OBES), Chemical Sciences, Geosciences and Biosciences Division, U.S. Department of Energy (DOE) under Contract No. AC03-76SF00098. Work at the University of Kentucky was supported by the National Science Foundation, Grant INT-9515504. XAFS data were collected at the Stanford Synchrotron Radiation Laboratory, which is operated by the DOE/OBES.

-
- ¹ L. D. Landau, *Sov. Phys. JETP* **3**, 920 (1957).
 - ² See, for example, articles in *Proceedings of the Conference on Non-Fermi Liquid Behavior in Metals*, Santa Barbara, California, 1996, edited by P. Coleman, M. B. Maple, and A. J. Millis, *J. Phys.: Condens. Matter* **8**, 9675 (1996).
 - ³ G. R. Stewart, *Rev. Mod. Phys.* **73**, 797 (2001).
 - ⁴ A. J. Millis, *Phys. Rev. B* **48**, 7183 (1993).
 - ⁵ L. B. Ioffe and A. J. Millis, *Phys. Rev. B* **51**, 16151 (1995).
 - ⁶ T. Moriya and T. Takimoto, *J. Phys. Soc. Jpn.* **64**, 960 (1995).
 - ⁷ G. G. Lonzarich, in *The Electron*, edited by M. Springford (Cambridge University Press, New York, 1997), chap. 6.
 - ⁸ P. Coleman, *Physica B* **259-261**, 353 (1999).
 - ⁹ J. A. Hertz, *Phys. Rev. B* **14**, 1165 (1976).
 - ¹⁰ P. Nozières and A. Blandin, *J. Phys. (France)* **41**, 193 (1980).
 - ¹¹ T. Graf, J. D. Thompson, M. F. Hundley, R. Movshovich, Z. Fisk, D. Mandrus, R. A. Fisher, and N. E. Phillips, *Phys. Rev. Lett.* **78**, 3769 (1997).
 - ¹² O. O. Bernal, D. E. MacLaughlin, H. G. Lukefahr, and B. Andraka, *Phys. Rev. Lett.* **75**, 2023 (1995).
 - ¹³ E. Miranda, V. Dobrosavljević, and G. Kotliar, *J. Phys. Condens. Matter* **8**, 9871 (1996).
 - ¹⁴ A. H. Castro Neto, G. Castilla, and B. A. Jones, *Phys. Rev. Lett.* **81**, 3531 (1998).
 - ¹⁵ A. H. Castro Neto and B. A. Jones, *Phys. Rev. B* **62**, 14975 (2000).
 - ¹⁶ E. Miranda and V. Dobrosavljević, *Phys. Rev. Lett.* **86**, 264 (2001).
 - ¹⁷ O. Trovarelli, C. Geibel, S. Mederle, C. Langhammer, F. M. Grosche, P. Gegenwart, M. Lang, G. Sparn, and F. Steglich, *Phys. Rev. Lett.* **85**, 626 (2000).
 - ¹⁸ F. M. Grosche, P. Agarwal, S. R. Julian, N. J. Wilson, R. K. W. Haselwimmer, S. J. S. Lister, N. D. Mathur, F. V. Carter, S. S. Saxena, and G. G. Lonzarich, *J. Phys. Cond. Matt.* **12**, L533 (2000).
 - ¹⁹ C. Petrovic, P. G. Pagliuso, M. F. Hundley, J. L. Sarrao, J. D. Thompson, and Z. Fisk, *J. Phys.:Condens. Matter* **13**, L337 (2001).
 - ²⁰ L. Shlyk, J. C. Waerenborough, P. Estrela, L. E. DeLong, A. de Visser, and M. Almeida, *J. Phys.: Condens. Matter* **11**, 3525 (1999), a displacement parameter $x=0.082$ was obtained, but not reported, in this study.
 - ²¹ A. H. Castro Neto (2002), private communication.
 - ²² L. Shlyk, P. Estrela, J. C. Waerenborough, L. E. DeLong, A. de Visser, D. P. Rojas, F. Gandra, and M. Almeida, *Physica B* **292**, 89 (2000).
 - ²³ D. L. Cox, *Phys. Rev. Lett.* **59**, 1240 (1987).
 - ²⁴ D. L. Cox and A. Zawadowski, *Adv. Phys.* **47**, 599 (1998).
 - ²⁵ M. Jarrell, H. Pang, D. L. Cox, and K. H. Luk, *Phys. Rev. Lett.* **77**, 1612 (1996).
 - ²⁶ M. Jarrell, H. Pang, D. L. Cox, and K. H. Luk, *Phys. Rev. Lett.* **78**, 1996 (1997).
 - ²⁷ P. Estrela, A. de Visser, F. R. de Boer, T. Naka, and L. Shlyk, *Phys. Rev. B* **63**, 212409 (2001).
 - ²⁸ D. L. Cox (2002), private communication.
 - ²⁹ J. S. Kim, J. Alwood, D. Mixson, P. Watts, and G. R. Stewart, *Phys. Rev. B* **66**, 134418 (2002).

- ³⁰ J. Bucher, P. G. Allen, N. M. Edelstein, D. K. Shuh, N. W. Madden, C. Cork, P. Luke, D. Pehl, and D. Malone, *Rev. Sci. Instrum.* **67** (1996).
- ³¹ C. H. Booth, Ph.D. thesis, Univ. of California, Santa Cruz (1996).
- ³² T. M. Hayes and J. B. Boyce, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1982), vol. 37, p. 173.
- ³³ G. G. Li, F. Bridges, and C. H. Booth, *Phys. Rev. B* **52**, 6332 (1995).
- ³⁴ See <http://lise.lbl.gov/R SXAP/>.
- ³⁵ E. D. Crozier, J. J. Rehr, and R. Ingalls, in *X-Ray Absorption: Principles, Applications, Techniques of EXAFS, SEXAFS, XANES*, edited by D. Konigsberger and R. Prins (Wiley, New York, 1988), p. 373.
- ³⁶ C. H. Booth, F. Bridges, G. H. Kwei, J. M. Lawrence, A. L. Cornelius, and J. J. Neumeier, *Phys. Rev. Lett.* **80**, 853 (1998).
- ³⁷ C. H. Booth, F. Bridges, G. H. Kwei, J. M. Lawrence, A. L. Cornelius, and J. J. Neumeier, *Phys. Rev. B* **57**, 10440 (1998).
- ³⁸ E. D. Bauer, C. H. Booth, G. H. Kwei, R. Chau, and M. B. Maple, *Phys. Rev. B* **65**, 245114 (2001).